Linear and Nonlinear Resonant Effects in Metallic Arrays of Sub-Wavelength Channels filled with GaAs

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ABSTRACT

We investigate on the interaction of surface plasmon modes with TEM, Fabry-Perot-like cavity modes in arrays of sub-wavelength slits filled with GaAs. A full control on the transmission process, which is mostly dictated by the geometrical parameters of the array, such as the slit length and width as well as the separation between the slits, is achieved and explained. The effects of the interaction of pure cavity modes and surface modes lead to the formation of an energy band gap, i.e. a spectral band where a drastic inhibition of transmission is induced by the coupling and back-radiation of the smooth-interface, unperturbed surface plasmon. Strong field localization in sub-wavelength regions boosts also the nonlinear response of the structure. The mere assumption that the metal is nonlinear via Coulomb and Lorentz contributions, and the introduction of high-index, nonlinear media, such as III-V semiconductors, in the sub-wavelength channels opens a cross-coupling of TE and TM polarizations for both pump and harmonic signals and makes it possible to generate both TE- and TM-polarized fields. These fields are generated even under high-absorption conditions, and survive thanks to a phase locking mechanism that sets in between the pump and its harmonics.

Keywords: Surface Plasmon, enhanced transmission, second harmonic generation, nonlinear interaction, down-conversion

1. INTRODUCTION

Since the first observation of enhanced optical transmission (EOT)[1], numerous efforts have been devoted to prove that strong field localization occur on the metal surface and inside the apertures under these circumstances[2-4]. These experimental demonstrations also suggested that a potentially strong nonlinear response may arise from the field enhancement in nano-structured metals, thus leading to enhanced harmonic signals. The generation of a second harmonic (SH) signal has been measured for a single aperture surrounded by grooves[5], for array of sub-wavelength holes of different shapes[6] and arranged in periodic or irregular patterns[5-9]. Also third harmonic generation (THG) has been demonstrated experimentally for a gold film patterned with nano-holes[10]. Due to their centro-symmetric nature metals do not have any intrinsic nonlinear term and harmonic generation in this context has been usually explained as the result of a symmetry breaking at the surface of the metal. Moreover the calculation of the generated signal has been always addressed by separating the nonlinear contributions into surface and volume sources, and by assigning to them suitable weights[11-14]. Another relevant feature in the nonlinear processes from sub-wavelength patterned metal is the nature of the aperture, i.e. the number of available conductors. Field localization and, as a consequence, harmonic generation is significantly different whether resonant modes are allowed inside the aperture (slits, annular structures) or not (holes). The ability of slits and annular structures to support TEM-like resonant modes[15-17] opens indeed other ways to generate harmonic fields when apertures are filled with nonlinear materials, such as $LiNbO_3$ or GaAs[18-20]: a significant improvement in second harmonic generated signal has been demonstrated for these structures when compared to bulk nonlinear material without the metal pattern.

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14. ABSTRACT
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The improvement of the nonlinear response is even more promising considering the thicknesses of the materials involved, very far from the coherence length of the nonlinear crystal. However, several aspects have been always ignored in these studies. We propose a study of harmonic generation from metal nano-patterned structures without imposing any separation between surface and volume sources, treating free electrons using the hydrodynamic model[21-25], making no a priori assumptions about charge or current distributions, and including Coulomb, Lorentz, convective, and linear and nonlinear contributions to the linear dielectric constant of the metal arising from bound (or valence) electrons[26]. In the analysis of harmonic generation from metals combined with nonlinear material[18-20] there is a tendency to focus only on nonlinear restoring forces and to neglect intrinsically nonlinear magnetic forces that drive all bound electrons, and to ignore harmonic generation arising from the metal itself. Indeed, while magnetic forces in bound electrons may be several orders of magnitude smaller than nonlinear restoring forces, they are always present and in fact play a catalytic role by activating new interaction channels among the different harmonics.

We considered a periodic arrangement of sub-wavelength slits milled on Ag and filled with a nonlinear material, i.e. GaAs. By considering a χ_2 tensor where the only non zero components are $d_{14} = d_{25} = d_{36}$, and modeling GaAs with all bound electrons, we calculate all the generated harmonic components (SH and TH both TE- and TM-polarized) and the down converted TE-polarized pump photons. The harmonic generation boosts further including third order nonlinearity of metal. Finally we clarify the role of phase locking process in the proposed structure. During the last few decades several groups have pointed out both theoretically and experimentally[27-33] the existence of a double peak structure in the SHG process under phase and group velocity mismatch conditions. This peculiar process remains valid also for negative index[33,34] or absorbing materials[35-37] thanks to a trapping and dragging mechanism between the fundamental and phase-locked generated pulse[32-37]. To corroborate the thesis that the phase locking of the pump and harmonic fields is not irrelevant in the structure under investigation, we tune the FF in the transparency region of GaAs and the SH and TH in a spectral range where the absorption is not negligible, so that the components that survive in the nonlinear medium are certainly propagating under the phase locking condition.

2. LINEAR RESPONSE OF A SILVER GRATING FILLED WITH GAAS

We begin our analysis by examining the behavior of a single slit of size *a* filled with GaAs, and carved on a silver layer[38] having thickness *w* (see Fig.1a). We tune the FF in a region of transparency (ε_{GaAs}(1064nm) ~ 12.10), while both second (532nm) and third harmonic (354nm) are tuned deep in the absorbing region (respectively ε_{GaAs}(532nm) ~ 17.08+ i2.86 and ε_{GaAs}(354nm)~8.81+ i14.36), where no harmonic generation is expected. As demonstrated theoretically and experimentally[5-10], strong field localization for the pump field supports second and third harmonic generation in properly dimensioned metal gratings. In order to favor nonlinear processes inside the nano-cavity one should optimize the linear transmission properties of the stack using incident TM-polarized light (electric field parallel to the y-axis in Fig. 1(a)). A single slit milled in a metal film supports TEM-like modes that exhibit field intensities as high as 100 times larger than the input field intensity[39,40].

In order to maximize the linear response at λ = 1064nm we varied the thickness of the silver film and aperture size and obtained a transmission map that reveals the strong resonant nature of the structure (Fig. 1(b)). Further enhancement of the linear response can be achieved by arranging the slit in a periodic pattern. The simulations were carried out on an infinite array of slits 60nm wide on a 100nm-thick silver film. The periodicity is varied from p = 200nm to p = 3200nm. For the sake of completeness, in Fig.2 we report the transmission response for an infinite array of slits for both TM (red line – square markers) and TE polarized (blue line – circle markers) fields. We note that the transmission is calculated by normalizing the outgoing energy to the energy that actually impinges on the geometrical area of the slits. As already pointed out elsewhere[41-44], the role of array periodicity (or pitch size) is detrimental to transmission if its value is a multiple of the surface plasmon wavelength of the dielectric/metal unperturbed interface, whose choice favors the opening of a plasmonic band gap. Moreover, slits have no cutoff for TM-polarized light, so that a TEM-like a resonant state is always available inside the slit for certain wavelengths and thicknesses. The interference of these horizontal resonances (light strongly confined along the x-axis) with the modes resonating in the vertical direction (surface waves along the y-axis) favors strong modulation of the linear transmission profile (Fig.2), causing the appearance of a gap every time the surface plasmon wavelength matches the periodicity of the array.

In the following section we will investigate the generation of harmonics of both polarizations. We will also discuss a novel down-conversion process that changes the polarization of incident TM-polarized pump photons into phase-locked TE-polarized light. Transmission values for an incident, TE-polarized pump field – Fig.2 – are less than 1% for large periodicities, and approach 1% when slit-to-slit-distance is relatively small. The reason for the enormous difference between the two polarizations is due to the fact that resonant Fabry-Perot modes are not accessible to TE-polarized light, which at 1064nm are well below the cut-off. We note that similarly to the TM-polarization case, TE-

polarized light also exhibits strong transmission minima due to the interference of horizontal and vertical resonances. However, while vertical modes for TM-polarized light can be ascribed to the coupling and back-radiation of surface plasmons on the impinging interface, these modes change their nature for a TE-polarized field, matching exactly the Rayleigh minimum condition.

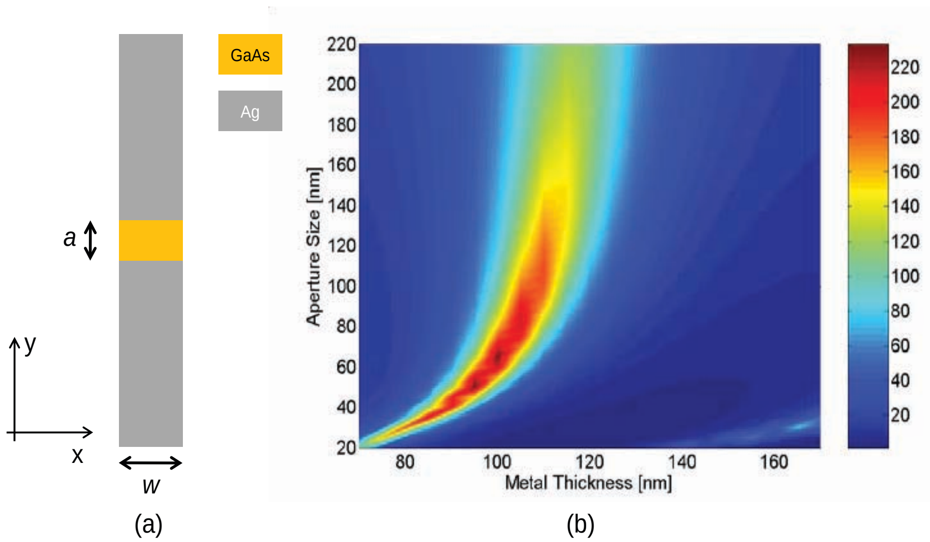

Fig.1. (a) Sketch of a single slit of size *a* filled with GaAs and milled in a silver film of thickness *w;* (b) Transmission map at λ=1064nm for a single slit carved on a silver substrate, filled with a material having ε_{GaAs}=12.10+ i0.

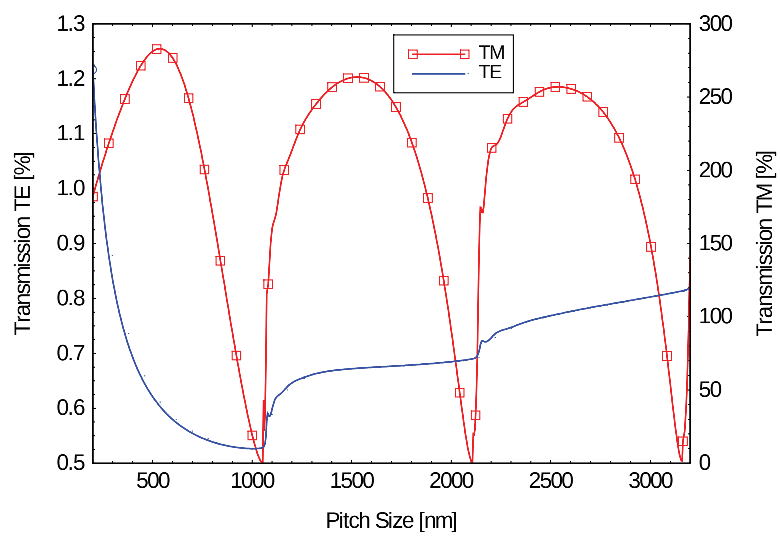

Fig.2. Transmission versus pitch size at 1064nm for both TM (red line – square markers, right axis) and TE (blue line – circle markers, left axis) polarization

3. NONLINEAR RESULTS

The enhanced transmission process at near-IR, visible and UV wavelengths, whether it is due to vertical or horizontal resonances or a combination of both, is always characterized by field localization, absorption and field penetration inside the metal because in these ranges transition metals display dielectric constants of order unity. The interaction of light with both free and bound electrons in metals becomes more efficient especially if the light is concentrated and

enhanced in small volumes, i.e. sub-wavelength slits. Moreover, when a material having non negligible $\chi^{(2)}$ and/or $\chi^{(3)}$ values fills the slits, new channels for harmonic generation become available and eventually lead to phase-locked pump photon down conversion.

Let us consider the same system described in Fig. 1(a), with 60 nm wide slits that are filled with GaAs and are arranged periodically on a 100 nm thick silver layer. The grating is illuminated with pulses approximately 120fs in duration, with peak intensities of roughly $2\mathrm{GW/cm^2}$. We calculated the nonlinear response considering bound and free electrons contribution arising from the metal, and bound electrons from GaAs, modeled as outlined in Ref. 26. For the sake of simplicity here we introduce quadratic and cubic nonlinear terms for GaAs only. The magnitude of the $\chi^{(2)}$ tensor of GaAs is chosen so that $2d_{14} = 2d_{25} = 2d_{36}$,=10pm/V, while $\chi^{(3)}$ is selected so that $\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy} = \chi^{(3)}_{zzzz} = 3\chi^{(3)}_{xxyy} = 3\chi^{(3)}_{xxzz} = 3\chi^{(3)}_{yyzz} = 3\chi^{(3)}_{zzyy} = 3\chi^{(3)}_{zzxx} = 3\chi^{(3)}_{xxyy} \sim 10^{-18}\,(\mathrm{m^2/V^2})$. As Figs. 3 and 4 demonstrate, an impinging TM-polarized field generates four nonlinear cross-polarized harmonic fields: TM-polarized SH and TH (Figs.3 (a) and (b) respectively), TE-polarized SH and TH (Figs.4 (a) and (b), respectively). If these results are read together with Fig.2 above, they reveal how the nonlinear response is dramatically influenced by the linear response for both polarizations: all the generated harmonics experience the same forbidden states as the incident pump field does. In addition, the harmonic are also constrained by the size of the wavelength relative to the geometrical characteristics of the structure. For example, the TM-polarized SH is strongly inhibited for pitch sizes matching the unperturbed air/silver surface plasmon wavelength of the pump and second harmonic. The same phenomenon is evident also for THG with appropriate pitch values. Note that for this assumed value of $\chi^{(2)}$ ~10pm/V the predicted conversion efficiency of the TM-polarized SH component (arising from the metal sections within the nanocavity) can be almost one order of magnitude larger than the TE-polarized SH conversion efficiency that arises from the GaAs itself.

One of our present objectives is also to demonstrate that the phase locking process briefly described above[27-33] is in fact playing a non trivial role in harmonic generation. A 100nm-thick GaAs substrate is only 20% transparent at 532nm, and completely opaque at 354nm. In a multi-pass geometry or a resonant nanocavity environment[37] the homogenous portion of the SH signal is removed more efficiently compared to bulk, so that all generated components that survive in the nonlinear medium are propagating mostly under the phase locking conditions. More convincing numerical evidence of phase locking may be achieved by increasing substrate thickness to ~170nm, and by reducing the width of the nano-channel down to 20nm, so that we are still operating under resonant conditions. The result is that conversion efficiencies do not vary significantly, even though all the TE-generated harmonics are now far below cut-off. This is a sure sign that phase locking is the main mechanisms that drives the harmonic field to resonate inside the cavity even if it is tuned to resonate at the pump frequency[37]. It is worth noting that the down-conversion to TE-polarized pump photons (see Fig.5) is not trivial because the transmission of an incident TE-polarized pump field in this structure should be completely forbidden, as waveguide theory suggests and Fig.2 demonstrates.

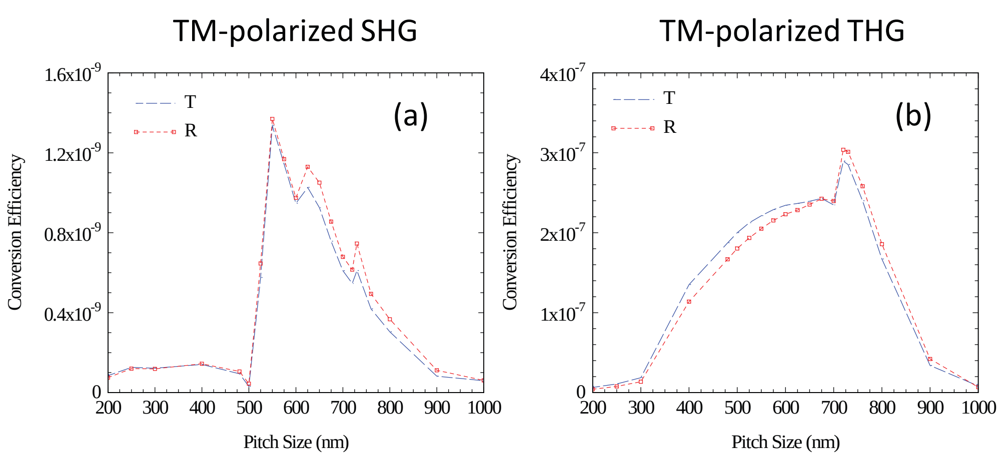

Fig.3: TM-polarized (a) second and (b) third harmonic transmitted (red line – square markers), reflected (green line – full circle markers) and total (blue line – empty circle markers) conversion efficiencies.

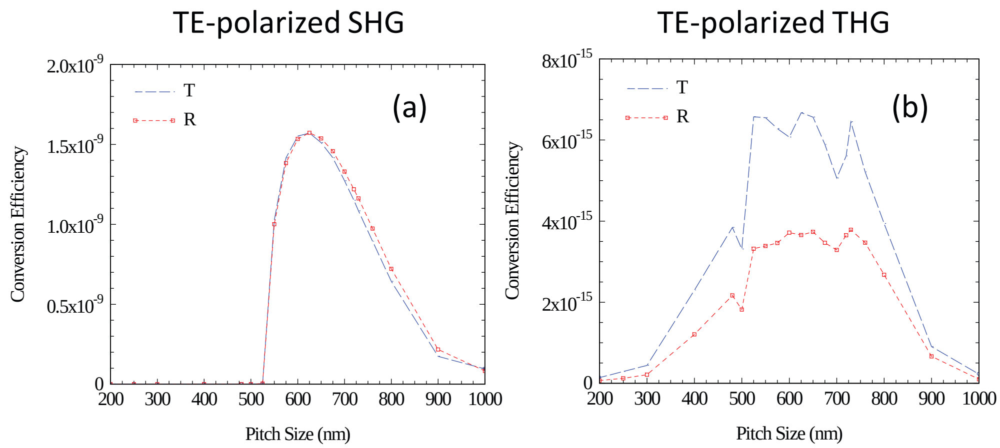

Fig.4: TE-polarized (a) second and (b) third harmonic transmitted (red line – square markers), reflected (green line – full circle markers) and total (blue line – empty circle markers) conversion efficiencies.

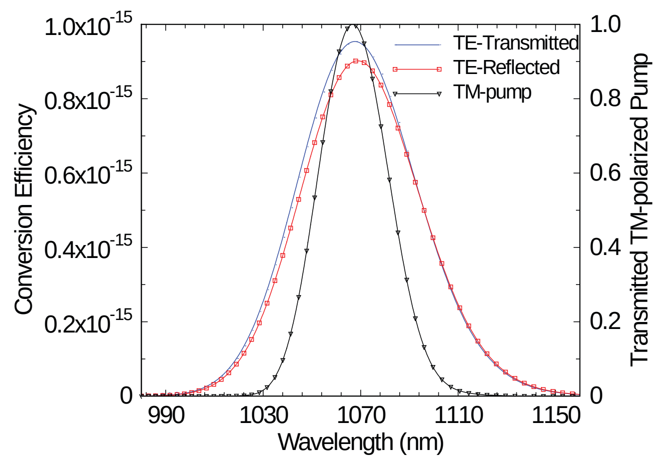

Fig.5: TE-polarized down-converted pump photon efficiency transmitted (blue line – circle markers) and reflected (red line – square markers) from an array of slits 60nm wide, filled with GaAs. The spectrum of the TE pump is compared with the incident TM pump (black line – triangle markers). Silver thickness is 100nm and array periodicity has been fixed to p= 590nm.

4. CONCLUSIONS

Second and third harmonic generation, as well as cross-polarized down conversion processes from GaAs filled sub-wavelength slits have been demonstrated using a general model[26] that allows to analyze linear and nonlinear dynamics without making any assumptions about either the roles or quantitative contribution of each type of nonlinear source, i.e. surface or volume terms. Harmonic generation in both polarizations has been shown to be possible thanks to the phase locking mechanism that takes place even in the enhanced transmission regime. The contribution of third order nonlinear component plays a relevant role in the dynamics of the whole system, boosting nonlinear features of the array. Further improvements are accessible through the introduction of nonlinear materials having diagonal nonlinear tensors.

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